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All-inkjet-printed MoS₂ field-effect transistors on paper for low-cost and flexible electronics

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Abstract

All-inkjet-printing of transistors has received much attention for low cost and flexible integrated circuits. However, most flexible field-effect transistors (FETs) based on the emerging two-dimensional materials suffer from the high cost of substrate and electrode materials. The requirements for high-temperature synthesis and precise control in processing add another layer of complexity. To overcome these issues, low-cost flexible paper-based MoS_2 FETs were fabricated by inkjet printing of MoS_2 channel materials on paper. Additionally, we proposed and achieved the mask-less and low-temperature formation of source and drain electrodes on paper using in-situ selective-area copper reduction. A low sub-threshold swing of 80 mV/dec, high on/off ratio of 10^5 , and very high turn-on current (I_{on}) of $200 \,\mu\text{A}$ of the paper-based flexible MoS_2 FETs were demonstrated using the proposed low-cost and facile all-inkjet-printing technique. The all-inkjet-printing technique assisted by in-situ copper reduction opens new opportunities for low-cost and batch fabrication of paper-based electronic devices in ambient conditions.

Keywords $MoS_2 \cdot Sub$ -threshold swing \cdot FETs \cdot Nanosheets

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Introduction

MoS₂ is one of the emerging materials for flexible electronics because of its unique tunable bandgap (direct bandgap $E_{\rm g} = 1.8$ eV for monolayer and indirect bandgap $E_g = 1.2$ eV for multilayer) and mechanical stability (Chung et al. 2014; Yao et al. 2013; Kim et al. 2016). Flexible MoS₂ field-effect transistors (FETs), photoelectric sensors, and logical switches were reported, showing its potential applications in wearable electronics (Alsaif et al. 2016; Kim et al. 2012; Lim et al. 2018; Das et al. 2013; Li et al. 2015a; Ziyu et al. 2018, 2019, 2020). Although few-layer MoS₂ was reported to exhibit a high current on/ off ratio of 1×10^8 and an electron mobility of 200 cm²/ (V s), the bottom-up approaches of chemical vapor deposition (CVD) or atomic layer deposition (ALD) of MoS₂ thin films often require transferring to high-κ dielectrics, and hence suffer from relatively low yield (Shin et al. 2018; Lee et al. 2015; Miyoshi et al. 2015). To improve the scalability and reduce cost, top-down methods, such as inkjet printing technology, were reported for the fabrication of FETs based on layered MoS₂ nanosheets (Castro et al. 2015; Li et al. 2016; Liu et al. 2015). MoS₂ inks



are obtained using liquid-phase sonication-assisted exfoliation. Few-layer, bilayer, and even monolayer MoS₂ nanosheets can be obtained. With intense research efforts on semiconductor inkjet printing, large-scale and low-cost all inkjet-printed flexible FETs have become a reliable and direct writing technology. Such dispersed nanosheets can form a device channel by continuous inkjet printing technology. Nanomaterials are widely used to develop novel environment friendly chemical mechanical polishing slurries (Zhenyu et al. 2012a, 2012b, 2013, 2018, 2020; Jing et al. 2019). However, interconnected networks based on gate-patterned FETs are still difficult to obtain with full self-alignment insulator-covered gate technology, nanoscale precision wafers and substrates are fabricated for the use in high performance devices of semiconductor and microelectronics industries. Such random MoS₂ nanosheets network channel has very limited mechanical resilience, breaking after hundreds of bending cycles (Bo et al. 2018; Zhenyu et al. 2015; Zhang et al. 2015). Therefore, reported flexible devices usually have low onstate current and poor sub-threshold swing (SS) when produced at large-scale, compared with traditional rigid ones (Shokouh et al. 2015; He et al. 2012; Zhenyu et al. 2017). Another challenge in the fabrication of flexible FETs is that the electrodes must be reliable in bending or folding tests. The mechanical adhesion between a metal electrode, e.g. silver, and a substrate, e.g. polyimide film, is poor (Li et al. 2014, 2015b; Castro et al. 2014; Akinwande et al. 2014). Additionally, silver electrodes were printed by expensive the Dimatix Materials Printer (DMP) 2000/3000 series inkjet printers, are not cost-effective. These studies are a breakthrough and milestone contribution compared with conventional industries, eliminating the pollution dramatically to the environment (Zhang et al. 2020; Cui et al. 2019; Junfeng et al. 2019).

In this paper, we propose and demonstrate a process that allows the effective and large scale production of all-inkjet-printed MoS₂ FETs on mechanically flexible paper substrates. Few-layer MoS₂ nanosheets, acting as the active channel of a FET, were printed with a conventional desktop printer (EPSON-LP310) on a pre-formed electrode array. The electrode array was made by in situ reduction of copper (Cu), i.e. selective area reduction at inkjet-printed areas. This method not only provides excellent mechanical adhesion of electrodes but also offers an effective means for low-cost fabrication of flexible electronics. Despite the paper surface roughness, the printed MoS₂ nanosheets can be firmly attached to the paper surface, which is beneficial for flex resistance. More importantly, a remarkable SS ranging from 80 to 90 mV/dec was demonstrated experimentally in back-gated FETs. The MoS₂ FETs show a high on/off ratio of 10⁵, a small

threshold voltage roll-off range from -1 to 1 V, and a high on-state current (I_{on}) at a low drive voltage V_{ds} of 3.0 V.

Results and discussion

We designed and implemented a process for the large-scale production of MoS₂ FETs using paper as the substrate and the gate dielectric. Figure 1a, b shows the sequential fabrication process of source/drain gate electrodes and channel layers of the all inkjet-printed MoS₂ FETs on paper. Initially, AgNO₃ precursor ink was deposited on the back of a piece of paper, and then printed into strip patterns by inkjet printing. An optical image of AgNO₃ precursor strips printed on paper is shown in Fig. 1c. After UV curing of the AgNO₃ precursor ink, in-situ Cu reduction was simultaneously conducted at the strip patterns and the back of the paper. The paper with cured AgNO₃ patterns was immersed in a Cu-containing catalyst solution. The source and drain electrodes were selectively formed at the pre-printed AgNO₃ strips by Cu reduction reaction in a Cu-containing catalyst solution at a low temperature of 60 °C, as illustrated in Fig. 1a. Figure 1d shows the Cu source/drain electrodes and the back-gate electrode deposited on the pristine paper. The Cu source/drain electrodes adopted exactly the shape of the AgNO3 precursor strips through the in-situ selectively area Cu reduction reaction. The MoS₂ ink (Fig. 1b and S1) was printed directly on Cu electrode arrays on the paper surface to form MoS₂ channel and thus one can greatly reduce the complexity in the flexible device fabrication. The MoS₂ ink was dried at 40 °C to remove ethanol and deionized water in polyvinyl pyrrolidone (PVP), and MoS2 channels with a length of 100 µm and widths of 100 µm and 1000 µm were formed. After the low-temperature annealing step, the all-ink-printed MoS₂ FETs were fabricated on paper, Fig. 1e shows the optical images of the fabricated transistors. Figure 1f shows the three-dimensional schematic of the paper-based MoS₂ FET. The effective channel length is controlled by the distance between the deposited Cu source and drain contacts.

Figure 2 shows the top-view and cross-sectional scanning electron microscopy (SEM) images of the MoS_2 transistors printed on paper. As shown in Fig. 2a, well-defined Cu electrodes are observed. The chemical composition of the deposited Cu electrodes was analyzed by energy-dispersive X-ray spectroscopy (EDS), as shown in Figure S2a, b. The back-gated MoS_2 FET printed on Cu electrode arrays was show confirmed by the EDS results. The Cu film also contains nickel and iron residues that originate from the Cu-containing catalyst solution. The gap between the two electrodes is smaller than the initial template made by $AgNO_3$ strip patterns (100 μ m). The reduced channel width is due to the invasion effect of $AgNO_3$ and swelling during Cu-reduction. The Cu films are compact with



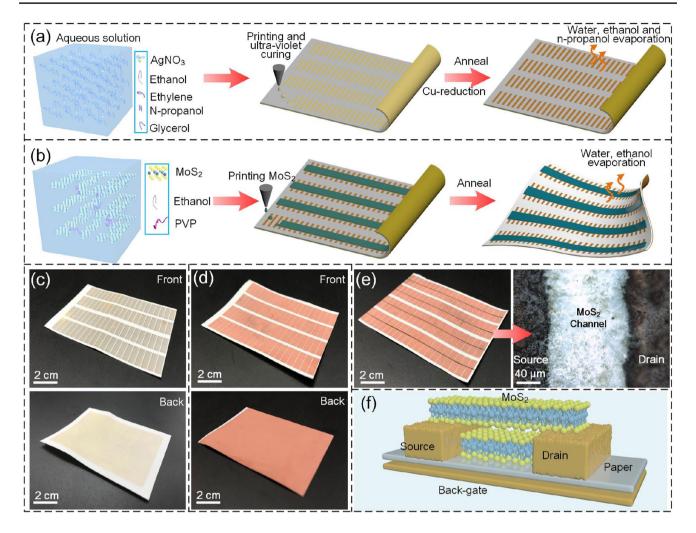


Fig. 1 The detailed fabrication process of the paper-based MoS_2 FETs. a $AgNO_3$ ink was prepared and printed on paper, the pristine $AgNO_3$ electrode patterns were obtained after ultra-violet curing, Cu electrodes arrays were then selectively reduced from the $AgNO_3$ patterns with HCHO catalyst at 60 °C. b Liquid-phase exfoliation to form few-layer MoS_2 nanosheet ink stabilized with PVP. The MoS_2 channel was printed between the Cu source/drain electrodes on paper,

and then the ${\rm MoS}_2$ channels was annealed at a temperature over 40 °C to remove deionized water and ethanol. **c** Optical images of printed ${\rm AgNO}_3$ seed patterns on both sides of the paper. **d** Optical images of Cu electrodes formed after selective reduction. **e** An optical image shows the fabricated ${\rm MoS}_2$ FET array printed on paper, and a magnified ${\rm MoS}_2$ FETs. **f** Schematic illustration of a ${\rm MoS}_2$ FET on paper substrate

small bumps, as shown in the SEM image of Fig. 2a. The film roughness be related to Cu can be explained by the formation of Cu clusters of different sizes as indicated in the inset of Fig. 2a. Most of these bulges could reduction at 60 °C. Figure 2b shows the SEM image of MoS₂ ink printed on the Cu electrodes. A compact MoS₂ film is formed over the electrodes with sharp interface, which show the potential to achieve narrow channel width. To investigate the quality of MoS₂ films deposited by inkjet printing, X-ray diffraction (XRD) measurements are conducted. The diffraction patterns dominated by the bulk MoS₂ (002) peak for films obtained at different exfoliation times, indicating that few-layer or bilayer MoS₂ nanosheets are restacked after drying. There are three diffraction peaks centered at 14.4°, 44.1°, and 60.1°, which are

indexed as (002), (006) and (008), respectively, as shown in Fig. S2b. It is confirmed that the stacked nanosheets of the two-hexagonal (2H) phase are randomly arranged during ultrasonic liquid-phase exfoliation. With ultrasonic exfoliation time increasing, the (100) peak at $2\theta = 34.3^{\circ}$ becomes increasingly apparent, and this observation shows that the PVP/MoS₂ ink consists of 2D MoS₂ nanosheets.

Figure 2c shows the cross-section morphology of the MoS_2 film. The low-magnification SEM image shows that the thickness of the MoS_2 film is rather uniform across a large area. The thickness of the printed MoS_2 channel is ca. 5 μ m as shown in Fig. 2d. Beneath the MoS_2 film, the Cu atoms are agglomerated as displayed in Fig. 2c, which may improve electron injection from the Fermi level of Cu contacts to the electron injection from the Fermi level



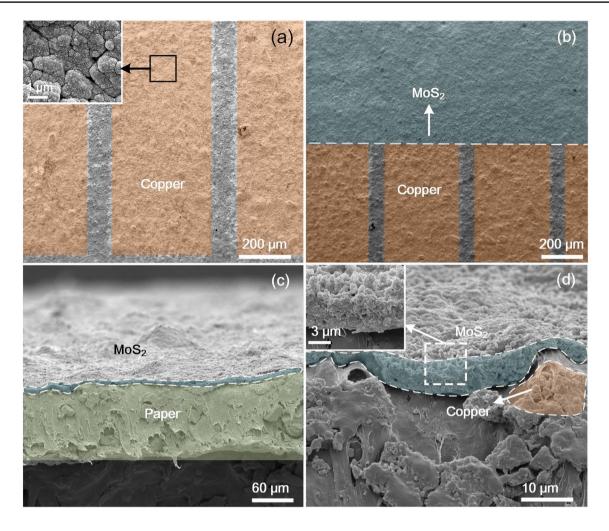


Fig. 2 SEM images of $\bf a$ the front of source/drain electrodes without printing MoS₂ active channel and $\bf b$ a 100 μ m MoS₂ channel length was printed on the paper, $\bf c$ SEM pictures of the surface of MoS₂ channel and $\bf d$ the contact between copper and MoS₂ channel

of Cu contacts to the conduction band of the conduction band of the MoS_2 channel.

Since the Cu source and drain electrodes are placed further apart (100 μm), it is necessary to evaluate MoS₂ nanosheets' size and ensure the interconnection between nanosheets in the device channel. The few-layer thickness of nanosheets is confirmed by transmission electron microscopy (TEM) images shown in Fig. 3. To demonstrate the sonication time effect on exfoliation, the pristine MoS₂ was sonicated for different times: 3 h, 24 h, and 48 h with PVP. Due to electrostatic repulsion, the surface energy of PVP, and van der Waals (vdWs) forces in MoS₂, these nanosheets are stable in the chemical solution. High-resolution TEM (HRTEM) was also employed to investigate the structural properties of MoS₂ nanosheets. Although the TEM measurements did not provide the layer number of MoS₂ nanosheets, the thickness of MoS₂ crystal can be indicated by its transparency.

Figure 3a shows a pristine MoS_2 crystal containing a large quantity of small MoS_2 nanosheets. The typical size of the MoS_2 crystal is about 4 μ m. A MoS_2 transistor based on the pristine MoS_2 crystal is rather poor in our tests, in terms of SS and on/off current ratio. After 3-h sonication, the size of a MoS_2 crystal is about 500 nm as shown in Fig. 3b. HRTEM image in Fig. 3c shows a number of small size multilayer MoS_2 nanosheets covering the MoS_2 crystal due to insufficient ultrasonic exfoliation. Although there are still some un-exfoliated MoS_2 crystals, the layered MoS_2 crystals have become increasingly thin.

After 24 h' exfoliation, as shown in Fig. 3d, the MoS₂ nanosheet thickness and size (*ca.* 300 nm) was further reduced. Figure 3e shows a magnified TEM image of a MoS₂ nanosheet. Multiple stacked MoS₂ layers are clearly observed. The inset of Fig. 3e shows the Fast Fourier transform (FFT) of the multilayer MoS₂, which shows a good single crystal structure. Compared with the MoS₂ sheets



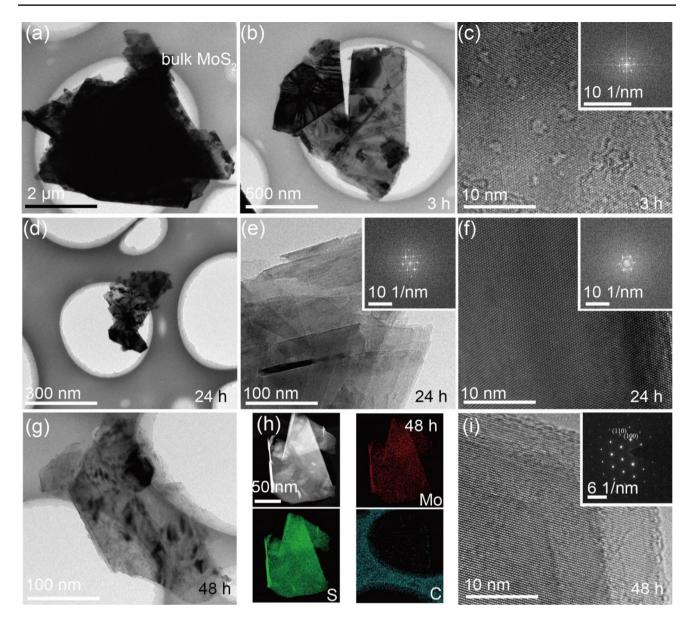


Fig. 3 TEM images of **a** the pristine bulk MoS₂ and **b** the exfoliated MoS₂/PVP crystals dispersed in ethanol after 3 h sonication at 70 W. **c** HRTEM image of MoS₂ nanosheets showing protuberant texture due to the overlap of small size MoS₂. **d** TEM image of MoS₂ crystals after exfoliation with ultrasound for 24 h. **e** The stack structure of MoS₂ nanosheets after 24 h sonication exfoliation and its corresponding Fast Fourier transform (FFT) shown in the inset. **f** HRTEM image

of few-layer MoS_2 nanosheets without multiple small-sized MoS_2 layers after 24 h' sonication exfoliation and the corresponding FFT pattern. **g** TEM image of few-layer MoS_2 nanosheet after 48 h sonication exfoliation. **h** TEM-EDS images of few-layer MoS_2 (48 h) and elemental mapping results on carbon framework. **i** HRTEM image of the selected region of few-layer MoS_2 nanosheets and its corresponding six-fold-symmetric diffraction spots SAED pattern

(3 h sonication) shown in Fig. 3c, small MoS₂ nanosheets stacked on a large size crystal do not appear, as shown in the HRTEM image of Fig. 3f. Defect-free lattice fringes of the MoS₂ nanosheets and FFT are clearly shown in Fig. 3f and the inset, respectively.

After 48-h sonication, a stable MoS_2 ink solution was obtained (Fig. S1). The TEM image in Fig. 3g illustrates the lateral sizes and the layered structure of nanosheets obtained after 48 h of ultrasonic exfoliation. The lateral size

was mainly distributed between 100 and 200 nm. The TEM-EDS elements mapping analysis of the MoS₂ nanosheets are presented in Fig. 3h. Elemental C originates from the ultrathin carbon-copper film substrate used for TEM analysis. Mo/S atomic ratio of few-layer MoS₂ nanosheets is about 1/2, which indicates ultrasonic exfoliation did not induce an unacceptably high defect concentration. This is a key observation since otherwise a highly defective MoS₂ channel increases the carrier scattering, compromising the



device performance. HRTEM image in Fig. 3i confirms the formation of a dispersive layer structure made of MoS_2 nanosheets. As the number of layers decreases, trilayer, and even bilayer MoS_2 nanosheets are possible. Few-layer MoS_2 crystals are critical for the formation of a continuous MoS_2 channel. The corresponding selected area electron diffraction (SAED) pattern is shown in the inset of Fig. 3i, confirming the hexagonal structure of MoS_2 and the diffraction spots for the (110) and (100) planes along the <100 > crystal axis.

The electrical characterizations, the I-V transfer and output characteristics of the MoS₂ FETs, were performed using a semiconductor analyzer (Agilent B1500a). The paper-based MoS₂ FET exhibits a typical n-type behavior, where the thickness of the oxide is kept constant at 100 μ m. The experimental and simulated Non-Equilibrium Green Functions (NEGF) results (Fig. 3s) are obtained varying the

source-to-drain bias (V_{ds}) from -2 to 2 V and back-gated $(V_{\rm gs})$ from 1 to 40 V in steps of 1.0 V, as shown in Fig. 4a. By taking optical phonon and acoustic phonon scattering into consideration, the simulated NEGF output currents agree with experimental values well. Therefore, the layered MoS₂ FET carrier transport probably presents multilayer optical phonon and acoustic phonon scattering. To evaluate the flexibility of our paper-based MoS₂ FETs, the MoS₂ FET array is bent at different angles under mechanical stress, as shown in Fig. 4b, c. A typical Ohmic contact behavior in the $I_{\rm ds}$ - $V_{\rm ds}$ output characteristics is observed at various $V_{\rm bg}$ after bending at 170° and 130°. Here, the output currents are the smallest for the largest bending angle and gradually decreased when increasing the bending angle. These results can be explained due to an increase of their respective contact resistance between the channel and source, so

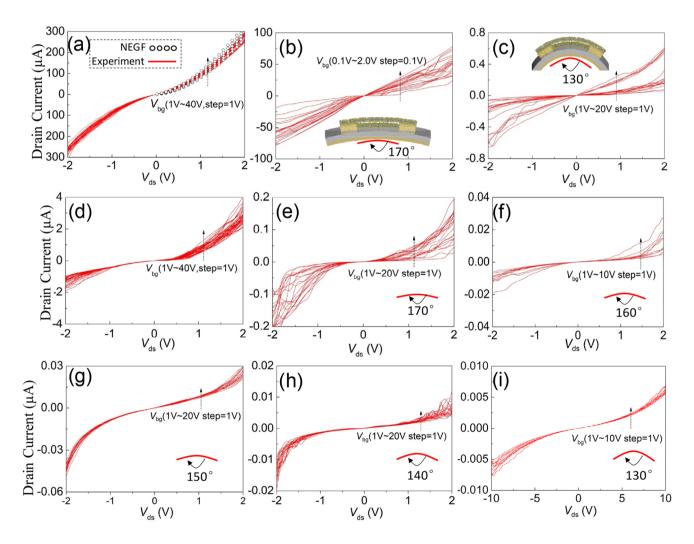


Fig. 4 Electrical properties of flexible Cu/MoS $_2$ contact FET under initial flat and bent condition. Output characteristics drain-to-source current versus drain-to-source voltage $(I_{\rm ds}-V_{\rm ds})$ curves under different gate bias $V_{\rm gs}$ show typical n-type gate dependence. MoS $_2$ FET with channel width $(W_{\rm g}=1000~\mu{\rm m})$ and length $(L_{\rm g}=100~\mu{\rm m})$ at vari-

ous bend angles, **a** the NEGF simulation results were compared with experimental results w/o bending ($V_{\rm bg}$ is in the range of 1–40 V), **b** bending angle is 170°, **c** 130°. MoS₂ FET with channel width ($W_{\rm g}=100~\mu{\rm m}$) and length ($L_{\rm g}=100~\mu{\rm m}$) at various bending angles **d** flat, **e** 170°, **f** 160°, **g** 150°, **h** 140°, and **i** 130°



carrier transmission coefficient decreases (Guo et al. 2014; Bartolomeo et al. 2018).

Figure 4d–i shows the output characteristic curves of the MoS_2 FETs with a narrow channel width, $W_g = 100 \, \mu m$, at different bending angles. Although the channel width reduced from 1000 to 100 μm , the output currents scaled down more than ten times due to the non-uniform distribution of MoS_2 nanosheets, and hence random interconnection networks in the channel. For the channel width of 100 μm , the I-V characteristics of MoS_2 FETs bending at 170° , 160° , 150° , 140° , and 130° are shown in Fig. 4e–i, respectively. The output current generally decreased with increasing bending angle. This current reduction is attributed to a random interconnecting network with reduced overlapping, and every transistor remains a constant degree after hundreds of bending. Table 1 shows the maximum on-state current at different bending angles, and the on-state current would

become weaken as the effective interconnection networks decrease.

To further elucidate the ${\rm MoS_2}$ FETs stability, the transfer curves from day 1 up to 30 days were analyzed. These results are presented in Fig. 5a for a ${\rm MoS_2}$ FET with $W_{\rm g}$ = 1000 ${\rm \mu m}$ and $L_{\rm g}$ = 1000 ${\rm \mu m}$ showing an obvious negative threshold voltage ($V_{\rm th}$) shift as the transistor ages. Threshold voltages are usually defined by the $I_{\rm ds}$ - $V_{\rm bg}$ characteristics by an abrupt current increase behavior in the sub-threshold region. It is worth noticing that the $V_{\rm th}$ shifted as the FETs aged, but the $I_{\rm on}/I_{\rm off}$ ratio is preserved to ~ 10⁴ and the SS ~ 80 mV/dec. The different $I_{\rm off}$ between Fig. 5a, b may originate from different interconnection network paths at the same drive voltage $V_{\rm ds}$. Additionally, the simulation NEGF results show different SS in the linear range. Since the NEGF calculated an integrated 2D ${\rm MoS_2}$ channel, but the experimental interconnection networks most likely contain several parallel

Table 1 Different device width of experimental results at various bending angles

Bending angle	$I_{\rm MAX}$, Flat (0°) at $V_{\rm ds} = 2 \text{ V}$	$I_{\rm MAX}$, 170° at $V_{\rm ds}$ = 2 V	I_{MAX} , 160°at $V_{\text{ds}} = 2 \text{ V}$	I_{MAX} , 150° at $V_{\text{ds}} = 2 \text{ V}$	$I_{\rm MAX}$, 140° at $V_{\rm ds}$ = 2 V	I_{MAX} , 130° at $V_{\text{ds}} = 2 \text{ V}$
Device size	'	,	,	,		
$W_{\rm g} = 1000 \; \mu \text{m}, L_{\rm g} = 100 \; \mu \text{m}$	210 μΑ	68 μΑ	X	x	X	$0.63~\mu A$
$W_g = 100 \ \mu \text{m}, \ L_g = 100 \ \mu \text{m}$	2.5 μΑ	57 nA	43 nA	28 nA	17 nA	5 nA

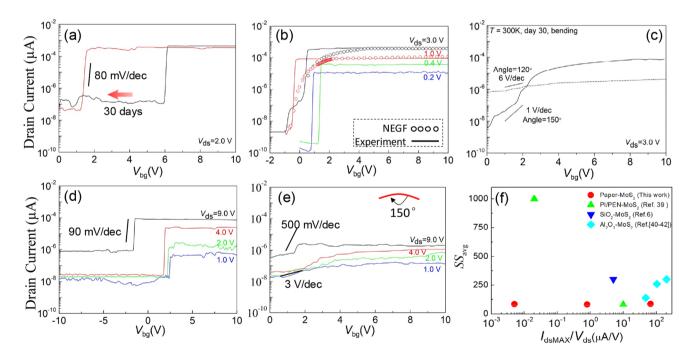


Fig. 5 The transfer characteristics of MoS $_2$ FET with $W_{\rm g} = 1000~\mu{\rm m}$ and $L_{\rm g} = 100~\mu{\rm m}$ of drain-to-sourcecurrent versus back-gated voltage $(I_{\rm ds} - V_{\rm bg})$ curves ${\bf a}$ at $V_{\rm ds} = 2.0~{\rm V}$ versus age after day 30 in air, showing a negative $V_{\rm th}$ shift, ${\bf b}$ NEGF and experimental results were investigated at different $V_{\rm ds}$ without bent after day 30 and ${\bf c}$ the trans-

fer characteristics under different bending angles (120° and 150°); The transfer characteristics of MoS₂ FET with $W_{\rm g}=100~\mu{\rm m}$ and $L_{\rm g}=100~\mu{\rm m}$ under different bending angle **d** flat, and **e** 150° at different $V_{\rm ds}$ after day 30. **f** A new method shows these performances of MoS₂-based on different substrates

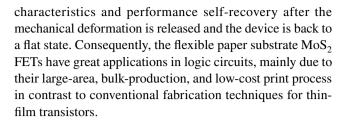


paths. Hence, with an increase in $V_{\rm bg}$, one of the interconnection network paths could firstly reach current saturation with small $V_{\rm ds}$. The variation of the $V_{\rm th}$ values is estimated to be shifted (from 1.2 to - 1.0 V) due to the random network path. As $V_{\rm ds}$ increases further, the interconnection networks will be significantly increased, which can be confirmed by the rapid increase of the saturation current at high $V_{\rm ds}$ (3.0 V). Figure 5c shows a comparison of the transfer characteristics of MoS $_2$ FET ($W_{\rm g}$ = 1000 μ m and $L_{\rm g}$ = 100 μ m) under different bending angle 120° and 150°. As the bending angle increases, the SS is far worse than the value of 80 mV/dec obtained without bending.

The transfer characteristics of MoS₂ FET ($W_g = 100 \mu m$ and $L_{\rm g} = 100~\mu{\rm m}$) showed $I_{\rm on}/I_{\rm off}$ ratio only ~ 10^2 and SS of 90 mV/dec at $V_{\rm ds} = 9.0$ V, as shown in Fig. 5d. The reduced on-state current could be attributed to a reduction of interconnection networks path. The threshold voltage for $V_{ds} = 1.0 \text{ V}$, 2.0 V, 4.0 V and 9.0 V were found to be 2.4 V, 2.2 V, 1.9 V, -2.0 V, respectively. After bending 150°, the SS becomes worse from 500 mV/dec to 3 V/dec at $V_{\rm ds} = 9.0$ V and 1.0 V respectively, as shown in Fig. 5e. As the $V_{\rm ds}$ increases further, the Schottky barrier height and carrier electron injection efficiency is reduced and improved, respectively, for larger bias voltage across the Cu-MoS₂ contact junction. Figure 5f shows the performance of devices made with the paper-based MoS₂ FETs. If symbol is near the right corner, the performance shall show good according to our new representation method. The outstanding electron mobility properties of MoS₂ on a large-area flexible PI substrate were illustrated (Akinwande et al. 2018). FETs were hand-made by ink-jetting MoS₂ inks on SiO₂ by a drop-casting process (Lim et al. 2018). MoS₂ nanoflakes are transferred on Al₂O₃-covered Si substrates (Shih et al. 2014; Hui et al. 2016; Jiahao et al. 2014), but the SS is also too high. Although FETs have undergone a process from simplicity to complexity, the all-inkjet-printed technology on paper substrates make MoS₂ flexible devices possible.

Conclusion

In summary, the flexible paper substrate n-type MoS_2 FETs on paper were fabricated and manufactured through the all-inkjet-printed process. The MoS_2 -based device on paper exhibits an on/off current ratio of 10^5 and also achieve good SS of 80 mV/dec with $W_\mathrm{g}=1000~\mathrm{\mu m}$ and $L_\mathrm{g}=100~\mathrm{\mu m}$ at $V_\mathrm{ds}=1.0~\mathrm{V}$, which is an important factor for application in high-speed circuits. We also discussed the carrier NEGF transport characteristics by matching to experimental results in a flat state. The transfer characteristics curves obtained at different bending angles show an influence of mechanical deformation in the electrical characteristics of the all-inkjet-printed FETs. However, MoS_2 FETs show current



Experimental section

Preparation of MoS₂ ink

MoS₂ powders are exfoliated in lift-assisted PVP and ethyl alcohol through bath sonication and centrifugation (7500 rpm) to obtain small MoS₂ nanosheets. Ethanol (20 ml) and deionized water (20 ml) were put in a sealed container mixed with PVP (80 mg), MoS₂ (440 mg), and sonicated at 27 °C for 48 h. The resultant suspension was centrifuged at 7500 rpm for 15 min to sediment thick nanosheets and the supernatant was obtained. Then, a stable MoS₂/PVP (2.4 mg/ml after evaporation of ethanol) dispersion was obtained.

Inkjet printing and electrode patterns

A redesigned commercial printer Epson-L310 was used to print the MoS₂ and AgNO₃. AgNO₃ ink was prepared with inorganic solvents, ethanol/water/ethylene glycol/npropanol/glycerol. AgNO3 electrodes pattern could be obtained via ultra-violet curing technique, the main concerns are the curing speed, and then Cu-containing catalyst solution consisted of CuSO₄·5H₂O, NiSO₄, K₄Fe(CN)₆, C₄H₄KNaO₆·4H₂O, C₁₀H₈N₂, and HCHO at an appropriate ratio that were injected to deionized water (10 g/L, 3 g/L, 0.04 g/L, 30 g/L, 0.02 g/L, 20 ml/L) in sequence, and the reaction of reduced Cu crystal particle is supposed to be the electrodes on surface of paper from Cu-containing catalyst solution with HCHO. MoS₂ channel layer was printed between the source/drain electrodes on paper by EPSON-L310. The deionized water and ethanol in the MoS₂ channel evaporate away over 40 °C for 10 min.

Characterization

Morphology of samples was observed by SEM (FEI NanoSEM50) Chemical identification of MoS₂ nanosheets was conducted by XRD (MAXima XRD-7000). Micro-level structure was characterized by TEM (FEI Tecnai G2F20 S-Twin). The direct current characteristics *I–V* curves of MoS₂ FETs were carried out using an Agilent parameter analyzer B1500a.



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Compliance with ethical standards

Conflict of interest There are no conflicts to declare.

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